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Emissions in combustion of lean methane-air and biomass-air mixtures supported by primary hot burned gas in a multi-stage gas turbine combustor

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Abstract

Thermal reaction of lean to ultra-lean premixed mixtures supported by the hot burned gas from the up-stream stage can be used for obtaining a better trade-off between ultra-low-NO_x and high combustion efficiency over a wide range of operations of a gas turbine. A three-stage model combustor designed based on this concept is being developed for a biomass gas-fueled regenerative cycle 10 kW micro-gas turbine. Tubular flame combustion is used for the primary stage and mixtures of lean to ultra-lean compositions are injected into the cross-flowing hot burned gas from the up-stream stage in the secondary and tertiary stages. The emissions and combustion characteristics are evaluated with methane and simulated biomass gas of different CO_2 contents at atmospheric pressure and inlet air temperatures up to 700 K.

In the experiments, the fuel flow for the secondary mixture was gradually increased while maintaining the fuel flow to the primary stage in the two-stage combustion mode and the fuel flow for the tertiary mixture was gradually increased while maintaining the fuel flows to the primary and secondary stages in the three-stage combustion mode. The combustor exit NO_x concentration corrected to 15% O_2 remained at or slightly lower than the level that was achieved at the start of fuel staging as far as the injected mixture was leaner than the primary mixture and NO_x emissions in the 10 ppm level were achieved at gas temperatures less than 1700 K. In contrast, the NO_x concentration increase steeply with equivalence ratio in non-staged combustion mode. The reaction of the injected mixture was completion when the reaction zone temperature was higher than 1500 K regardless of inlet air temperature and equivalence ratio of the primary stage. The results show that, the multi-stage combustion where mixtures of ultra-lean to lean compositions are injected into the hot burned gas from the up-stream stage achieved low-NO_x emissions and high combustion efficiency over a wide range of overall equivalence ratios or combustor exit gas temperatures. It is found that the CO_2 in the simulated biomass gas suppresses the NO formation by slowing down the progress of combustion reaction, especially at high temperature conditions.

Keywords: Low-NO_x emissions; Multi-stage combustion: micro-gas turbines; Biomass gas; Flameless combustion

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1. Introduction

Distributed co-generation using micro-gas turbines (MGT) has received a lot of attention as a means of efficient utilization of energy and protection of atmospheric environment. Biomass gas is renewable in nature and its combustion does not contribute to the net atmospheric concentration of carbon dioxide, a major green house gas. Therefore, the use of biomass gas in gas turbines in distributed co-generation systems should be promoted.

A project for developing a biomass gas-fueled regenerative cycle MGT of 10 kW output is being conducted in the Department of Mechanical Engineering, the University of Tokyo [1]. A multi-stage low-NO_x combustor for the MGT is being developed in collaboration with Aeroengine Environment Technology Center, JAXA. The air flow rate and operating pressure of the MGT are 194 g/s and 0.27 MPa and the combustor inlet air and exit gas temperatures are 700 and 1100 K, respectively, at the rated conditions. Emissions of NO_x less than 10 ppm (15% O_2) and combustion efficiency greater than 99.8% over the most part of and 99% over the whole operating range of the MGT.

The heating value of biomass gas is low and the compositions and hence heating values fluctuate depending on the methods of gasification and the compositions of raw biomass. These pose problems to the use of biomass gas as a fuel for gas turbines and, therefore, simultaneous achievement of high combustion efficiency and ultra-low-NO_x emissions would be more difficult.

Flameless combustion or thermal reaction [2,3] seems to be an approach to cope with the problems and to achieve the targets. An application of flameless combustion of fresh mixtures mixed with internally recirculated hot burned gas is investigated in a reverse-flow gas turbine combustor arrangement [4]. It is difficult to recirculate a sufficient amount of burned gas to mix with fresh mixture without deteriorating flame stabilization in single-stage combustion. In the present study, an axially multi-stage combustor configuration where ultra-lean to lean mixtures are injected into the hot burned gas from the up-stream stage [5,6] is being developed since it is easy to mix fresh mixture with required amount of hot burned gas without affecting the flame stabilization in the up-stream stage. This concept was successfully demonstrated in a lean–lean premixed pre-vaporized combustor for a 300 kW regenerative cycle gas turbine [7,8].

A three-stage model combustor is being developed for the MGT. This paper describes the emissions and combustion characteristics of the combustor operated at atmospheric pressure using pure methane and simulated biomass gas.

2. Model combustor

A schematic drawing of the model combustor is shown in Fig. 1. The major dimensions of the combustion liner are given in the figure. The combustor consists of three premixed combustion stages followed by a dilution zone. No liner cooling air is admitted. Good flame stability at lean fuel compositions is particularly required for the primary stage in order to achieving ultra-low-NO_x emissions fuel since the amount of NO_x formed in the primary stage often dominates the

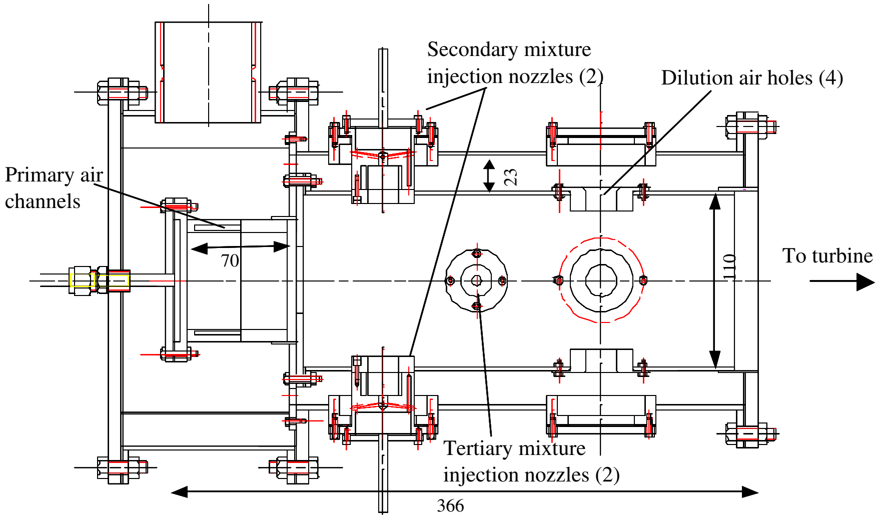


Fig. 1. Schematic drawing of model combustor.

combustor exit NO_x level [5,6]. In view of the flame stability, tubular flame combustion [8,9] is selected for the first stage. In the second and third stages, thermal reactions (or flameless combustion) of lean to ultra-lean mixtures injected into the cross-flowing hot burned gas from the upstream combustion stage is used to achieve high combustion efficiency while suppressing NO formation.

A pair of mixture injection nozzles, opposing each other, are provided for injecting mixtures into the secondary and tertiary stages. Schematic drawings (left) and photographs (right) of the mixture injection tube are shown in Fig. 2. The inner and outer diameters and length of the nozzle are 20, 40, and 55 mm, respectively. Eight curved narrow channels, 2.3 mm in width and 15 mm in height, are provided on the cylinder wall for imparting swirl motion to the mixture jet from the nozzle. Fuel gas is injected from the multiple fuel holes into the air flowing through the curved channels. This swirl motion enhances mixing of the injected mixture jet with the burned gas in the combustion section. Eight air holes, 2.3 mm in diameter, are drilled in radial direction through the wall of the cylinder to introduce air to the center hole, 6.5 mm in diameter, of the nozzle. Fuel gas is injected from the multiple fuel holes into the air flowing through the air holes and the resulting mixture goes out along the center line of the nozzle. Preliminary experiment showed that the injection of mixture from the center hole is very effective in suppressing the formation of recirculating flow and, therefore, preventing flashbacks into the nozzle.

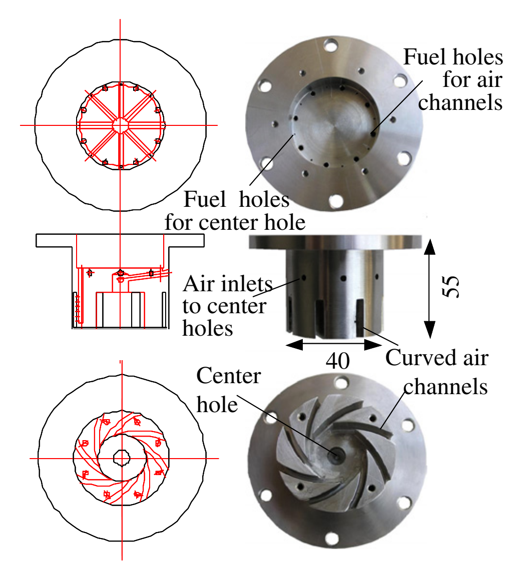


Fig. 2. Schematic drawing (left) and photo (right) of mixture injection nozzle.

3. Experimental procedure

All experiments in the present study were conducted at atmospheric pressure. The air to the combustor was pre-heated by an electric heater and its flow rate was measured by a vortex mass flow meter before pre-heating. The fuel gas was supplied from gas cylinders with supplying pressure adjusted by a regulator. The fuel flow to each combustion stage was controlled and measured by a mass flow controller. The inlet air temperature, T_{in}, was measured by a K-type thermocouple placed at the entrance of the combustor. The combustor pressure loss, ΔP, defined as the difference between the pressure measured at the combustor inlet and atmospheric pressure, was measured by a capacitance type pressure transducer.

The air flow rate was set at 72 g/s throughout the present study to establish the reference velocity in the combustor at the design inlet air temperature of 700 K though experiments were also conducted at 323, 500, and 600 K. The air splits between primary, secondary and tertiary stages and dilution zone were estimated to be 0.157, 0.177, 0.177, and 0.489 based on the measurements of flows to each stage.

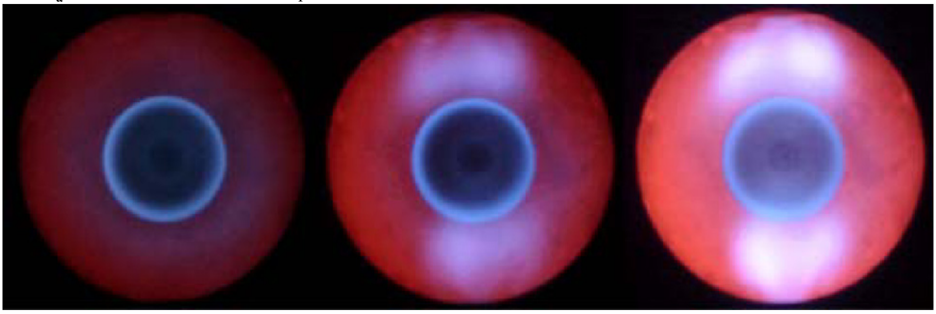
Gas sampling was made at the combustor exit with an X-shaped, water-cooled gas sampling probe with 32 holes. The compositions of gaseous species were determined by the standard gas analysis procedures. Chemiluminescence detection was used for NO, paramagnetic pressure difference was for O_2, non-dispersed infrared absorption was for CO and CO_2, and flame ionization detection was for HC (as CH_4). A catalytic converter, with verified conversion efficiencies greater than 95%, was used to convert NO_2 into NO for the determination of NO_x. Concentration of NO_x corrected to 15% O_2 and combustion efficiency were calculated from these data.

4. Experimental results

4.1. Observation of secondary mixture jets (Flameless combustion)

A digital camera was used to record the phenomena in the combustion chamber through the quartz window facing the combustion liner dome. Figure 3 shows typical photos recorded when ultra-lean to lean premixed secondary mixtures were injected into the hot burned gas from the primary stage. The gas temperature was estimated as 1230 K. The tubular blue flame in the primary stage is clearly seen and the stepped wall is heated in red. A pair of luminous reaction volumes are seen to develop from the exits of the mixture injecting nozzles at $\phi_2 = 0.6$ and faint bluish reaction volume are located at a distance from the exits of the mixture injecting nozzles at $\phi_2 = 0.4$.

$W_a = 72$ g/s Tin = 700 K $\phi_1 = 0.46$ (1230 K)



ϕ_2 =	0.25	0.4	0.6
Combustion efficinecy (%) =	99.9	99.9	99.9
NOx ppm (15 %O_2) =	4.9	7.1	35.6

Fig. 3. Photo of phenomena in combustor with secondary mixtures of different equivalence ratios injected.

No visible volumes are seen despite complete reaction at $\phi_2 = 0.25$. Ultra-lean mixture is reacted in flameless combustion or thermal reaction mode. It should be noted that the visible reaction volumes were never self-sustained. They were instantaneously blown off when the primary flame was extinguished by shutting off the fuel flow. Therefore, it was confirmed that the secondary stage combustion was supported by the hot burned gas from the primary stage.

4.2. NO_x emissions and combustion efficiency

Figure 4a shows the NO_x emissions corrected to 15% O_2 and combustion efficiency at an inlet air temperature of 700 K as a function of overall equivalence ratio, ϕ_t, determined by gas analysis. The equivalence ratio by gas analysis agreed with that from the measured air and fuel flow rates within an error of 3%. The circles show the data in the single-stage combustion mode where the fuel flow to the primary stage was varied. The squares show the data in the two-stage combustion mode where the fuel flow to the secondary stage was varied. The primary zone equivalence ratio was set at 0.46, where combustion of the primary mixture was complete, and no fuel was injected into the tertiary stage. The triangles show the data in the three-stage combustion mode where the tertiary fuel flow rate was varied while maintaining the primary and secondary mixture equivalence ratios at 0.46 and 0.43, respectively. The secondary mixture reacted completely at 0.43. The solid and open symbols represent the NO_x emissions and combustion efficiency, respectively.

In the single-stage combustion mode, the NO_x emissions level increases very steeply with overall equivalence ratio, ϕ_t. This steep increase implies good mixing of fuel and air because it reflects the well-known very strong dependence of the rate of thermal NO formation on equivalence ratio in premixed combustion. Low-NO_x emissions (less than 10 ppm) and high combustion efficiencies (greater than 99%) is achieved within a narrowly limited range around $\phi_t = 0.07$.

In the two-stage combustion, the combustion becomes completion after a small drop due to incomplete combustion with increasing the secondary fuel flow. The compatibility of low-NO_x emissions with high-combustion efficiency is achieved over the 0.11–0.15 overall equivalence ratio range. However, the NO_x level begins to increase very steeply when ϕ_2 exceeds about 0.5.

In the three-stage combustion, the range of equivalence ratios where both low-NO_x emissions and high combustion efficiency (greater than 99%) expands to 0.11–0.23, with achieving combustion efficiency greater than 99.9% over the most of the range. The NO_x level begins to increase very steeply when ϕ_3 exceeds about 0.5.

The drop in combustion efficiency occurs at the beginning of the tertiary mixture injection is much less than that at the beginning of the secondary mixture injection, as shown in Fig 4a. The difference is attributed to the fact that the drop in gas temperature due to the introduction of fresh mixture in the tertiary stage is smaller than that in the secondary stage. The amount of burned gas to the tertiary stage was about twice of that to the secondary stage though the amounts of fresh mixture to both stages were the same. A better trade-off between low-NO_x emissions and high combustion efficiency can be more easily obtained with increasing the number of combustion stages.

A comparison of the data in two- and three-stage combustion modes at $\phi_t = 0.2$ (near the design condition of the MGT) shows that the NO_x emission in the three-stage combustion mode is one-tenth of that in the two-stage combustion though combustion efficiencies greater than

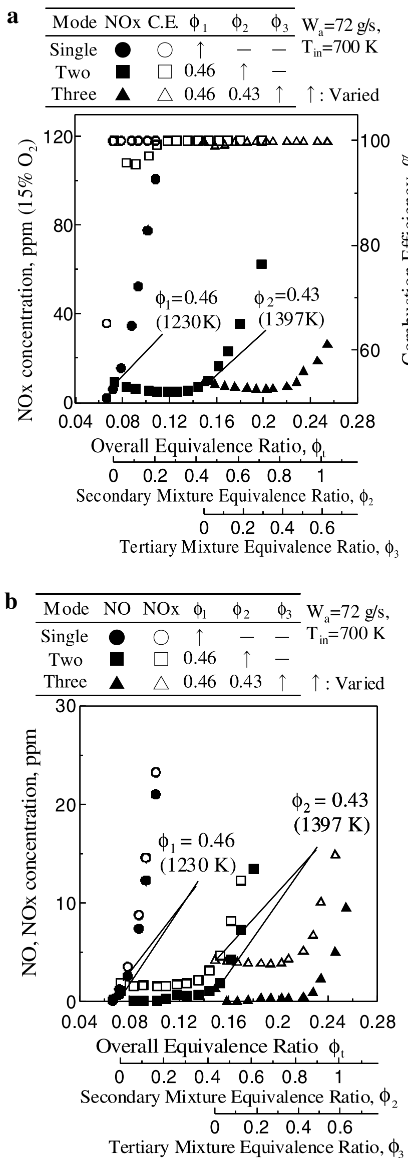


Fig. 4. NO_x emissions and combustion efficiency in the single- and multi-stage combustion; (a) NO_x concentration corrected to 15% O_2; (b) Raw concentration of NO and NO_x.

99.9% were achieved both in two- and three-stage combustion modes. It is obvious that both low-NO_x emission and high combustion efficiency were achieved by three-stage combustion at the rated conditions.

In supplementary experiments at 323, 500, and 600 K inlet air temperatures, the equivalence ratios of the primary and secondary mixtures were adjusted so that these mixtures could be completely reacted before entering the next stage, as was the case with the experiments at 700 K. The data at these inlet air temperatures are not shown here but the variations of NO_x emission and combustion efficiency with equivalence ratio are quite similar to those shown in Fig. 4a. This fact suggests that both low-NO_x emissions and high combustion efficiency can be obtained over the whole range of operations of the MGT.

Figure 4b shows the raw concentrations of NO_x and NO as a function of ϕ_t. The NO in the burned gas from the up-stream stage seems to be completely converted into NO_2 by the fuel in the secondary or tertiary mixtures. NO formation is almost zero for ultra-lean to lean mixtures (ϕ_1, $\phi_2 < 0.4$). The steep increases in NO_x concentration with increasing the equivalence ratio of the secondary or tertiary mixture beyond 0.4 is attributed to the formation of NO.

Severe combustion oscillation occured in the single-stage combustion mode, when the primary mixture equivalence ratio exceeded 0.8. In contrast, the operation in the multi-stage combustion mode was absolutely free from oscillation. This shows that the axially staging of lean to ultra-lean mixtures is a very promising approach to avoid combustion oscillation.

4.3. Carbon monoxide and hydrocarbons emissions

Figure 5 shows the raw, non-corrected, concentrations of CO and HC (as CH4) in the single-, two-, and three-stage combustion as a function of ϕ_t. Generally, the features of the variations of CO and HC emissions with ϕ_t are quite similar. In the single-stage combustion, both emissions are decreasing monotonically with increasing ϕ_t or ϕ_1 and become low enough at ϕ_t

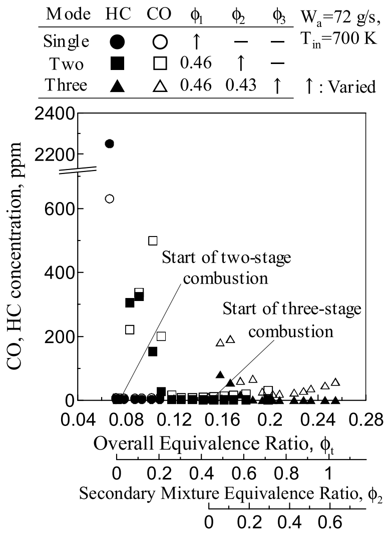


Fig. 5. CO and HC emissions in the single- and multi-stage combustion.

greater than 0.07 ($\phi_1 = 0.46$). In the two- and three-stage combustion, they increase to their peaks and then decrease towards zero as the secondary or tertiary fuel flow increases. They become low enough at ϕ_t greater than 0.12 ($\phi_2 = 0.25$) in the two-stage combustion and at ϕ_t greater 0.19 ($\phi_3 = 0.25$) in the three-stage combustion. A comparison of the variations of CO and HC emissions with ϕ_t shows conversion of HC into CO is enhanced gas at higher temperatures.

The CO and HC emissions are increasing with secondary or tertiary fuel flow rate when the secondary or tertiary mixture is too lean to react at a reasonably faster rate in the each combustion zone. A further increase in the fuel supply finally results in an increase in the reaction rate due to the increased reaction zone gas temperature, resulting in a rapid destruction of raw fuel into HC, HC to CO, and CO to CO_2. The peaks of CO and HC in the three-stage combustion are significantly lower than the counter parts in the two-stage combustion in spite of the fact that mixtures of similar equivalence ratios are injected into the secondary and tertiary stages. It is attributed to the fact that the reaction zone gas temperature in the former is higher than that in the latter.

4.4. Effects of primary mixture equivalence ratios

The effects of the primary mixture equivalence ratio, ϕ_1, on the NO_x emissions and combustion efficiency in the two-stage combustion mode are shown in Fig. 6 by comparing the data for $\phi_1 = 0.4$, 0.46 (already shown in Fig. 4) and 0.5. Combustion of the primary mixture is complete for $\phi_1 = 0.46$ and 0.5 but far incomplete for 0.4.

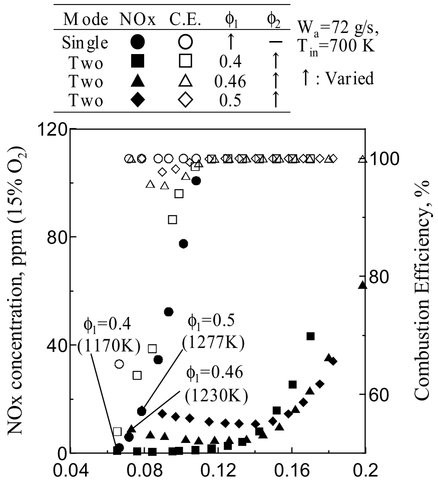


Fig. 6. NO_x emissions and combustion efficiency for different primary equivalence ratios, ϕ_1, in the two-stage combustion.

In the case of $\phi_1 = 0.4$, the combustion efficiency monotonically increases from about 50% toward 100% with increasing ϕ_2 and become near completion when ϕ_2 reaches 0.25. Introduction of ultra-lean secondary mixtures into the completely reacted burnt gas from the primary stage results in about 2% and 5% drops in combustion efficiency for $\phi_1 = 0.46$ and 0.5, respectively. The initial temperature of the secondary mixture after mixed with the hot burned gas from the primary stage is a critical factor for the initiation of the secondary mixture reactions, especially in flameless combustion mode. Complete reaction can be established if the initial mixture temperature is above the threshold. Assuming complete mixing of the secondary mixture and primary hot burned gas, the initial temperature in the secondary stage is estimated as 1170, 1230, and 1277 K for $\phi_1 = 0.4$, 0.46, and 0.5, respectively.

The NO_x emissions level corrected to 15% O_2 are once gradually decreasing from the level achieved with no secondary fuel injection to a minimum and then begins to increase steeply with increasing the secondary fuel flow rate for $\phi_1 = 0.46$ and 0.5. This decreasing trend is due to the fact that NO is not formed at all by the reactions of ultra-lean to lean mixtures ($\phi_1 < 0.4$) mixed with hot burned gas and the increasing trend is attributed to thermal NO formation.

4.5. Combustor exit NO_x emissions vs. theoretical gas temperature in each combustion stage

Figure 7 shows the combustion efficiency and NO_x emissions for different inlet air temperatures as a function of theoretical gas temperature in each combustion stage. It is assumed that the

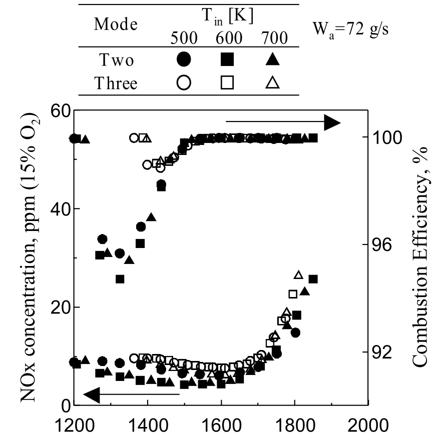


Fig. 7. NO_x emissions and combustion efficiency in secondary and tertiary combustion zones for different inlet air temperatures, T_{in}.

burned gas from the up-stream stage and the injected mixtures mix homogeneously and react completely. It is found that both combustion efficiency and NO_x emissions data for different inlet air temperatures are well correlated by the gas temperature in the stage.

Generally, the manner for the combustion efficiency to approach 100% with gas temperature is independent of the number of combustion stages and inlet air temperature. The combustion efficiency reaches 99% at a gas temperature of around 1500 K and 99.9% at around 1600 K. The drop in the combustion efficiency curve is shallower for the three-stage combustion than for the two-stage combustion. It is attributed to the higher gas temperatures in the tertiary stage than in the secondary stage.

The manner for the NO_x emissions to decrease gradually to a minimum and then to begin to increase steeply with increasing gas temperature is almost independent of the number of combustion stages and inlet air temperature. A much better correlation of the NO_x emissions with gas temperature is obtained for the data in the three-stage combustion as compared with the data in the two-stage combustion. The NO_x emissions levels at gas temperatures lower than about 1500 K scattered since the actual gas temperatures should be lower depending on the degree of incomplete combustion than the theoretical one. The minimum NO_x emissions in the order of 10 ppm occurr at around 1700 K gas temperature, above which NO_x formation via thermal mechanisms is enhanced. It is concluded from the above discussion that the splits of air and fuel between stages should be optimized so that the stage gas temperature is between 1500 and 1700 K in designing ultra-low emissions, multi-stage combustors.

4.6. Emissions with simulated biomass gas

The composition and hence heating value of digestion gas, one of the major biomass fuels, fluctuate, as previously mentioned. The effects of CO_2 content and heating values on the emissions and combustion characteristics of the combustor were investigated by using mixtures of methane and carbon dioxide of different CO_2 contents (25%, 40%, and 50%) were used to investigate The calorific values ranged from about a half to three forth of that of pure methane.

Figure 8 shows the NO_x emissions and combustion efficiency in the two-stage combustion mode. The primary stage was operated with methane at $\phi_1 = 0.46$ and the secondary stage with the synthetic biomass gas. No effects of CO_2 content on combustion efficiency are seen over the whole range of ϕ_2 tested. On the other hand, the effects of CO_2 content on the NO_x emissions become increasingly appreciable when ϕ_2 exceeds 0.5. An

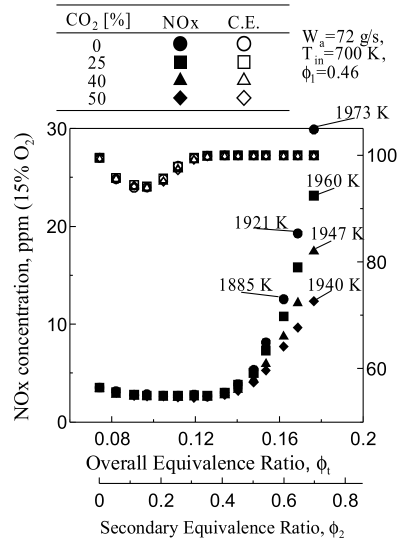


Fig. 8. NO_x emissions and combustion efficiency for simulated biomass gas of different CO_2 contents as secondary fuel (primary fuel: methane).

increase of CO_2 content reduces flame temperature in the secondary stage and, therefore, the thermal NO formation proceeds at a slower rate. A plot of the NO_x concentrations against the calculated flame temperature has revealed that, at a fixed gas temperature, the NO_x concentration was lower for higher CO_2 contents. The observed chemical effects are consistent with chemical kinetics calculation reported previously [10,11]. Preliminary kinetic calculations with CHEMKIN, using GRI-mech 3.0, suggest that CO_2 in the reactive gas slowdown the reactions and then reduced the emissions of NO via the thermal NO mechanism by shortening the effective residence time.

5. Summary

The emissions and combustion characteristics of three-stage micro-gas turbine combustor were investigated at atmospheric pressure and inlet air temperatures up to 700 K with methane and simulated biomass gas of different CO_2 contents. Tubular flame combustion was used for the primary stage and mixtures of lean to ultra-lean compositions were injected into the cross-flowing hot burned gas from the up-stream stage in the secondary and tertiary stages. In the thermal reaction mode, mixtures of ultra and very lean compositions reacted completely without forming NO. Most of NO produced in the up-stream stage was converted to NO_2 by the fuel in the secondary or tertiary mixtures. The gas temperatures of 1500 and 1700 K were the thresholds for combustion efficiency greater than 98% and for 10 ppm level

NO_x emissions, respectively. The CO_2 in the simulated biomass gas suppressed the NO formation by slowing down the progress of combustion reaction, especially at high temperature conditions. No combustion oscillation occurred in the stage combustion.

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Comment

Peter Jansohn, Paul Scherrer Institute, Switzerland. Did you need to adjust your fuel nozzle geometry due to the higher volumetric fuel flow rate in the case of CO_2-diluted fuel mixtures? What are the effects on the flow L mixing field?

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